

Fabrication of Multi-Bit Memory Devices Based on Layered Semiconductors *via* Interlayer Deformation

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The cutting-edge technology areas, such as hardware-level artificial intelligence (AI) machines, fly-by-feel control systems for next-generation vehicles, and neuromorphic ICs for advanced learning machines, need novel solid-state memory solutions with a lower cost-per-bit, a better scalability in comparison with the state-of-the-art memory devices, and new logic components with multiple (or analog-tunable) levels of excitation (or data storage) states.^[1-2] Several prototype devices have been fabricated and tested to realize multibit (or analog) memory states. For example, Shen *et al.* demonstrated neuromorphic modules based on randomly dispersed carbon nanotubes (CNTs).^[3-4] Lu *et al.* fabricated synaptic memory systems based on memristor devices.^[5] Additional upscalable nanofabrication methods capable of generating reliable analog memory states in electronic materials are highly desirable.

Here, we report that the nanoimprint-assisted shear exfoliation process can generate interlayer deformation features in exfoliated layered semiconductor (*e.g.*, transition metal dichalcogenides (TMDCs)) structures. Such deformation features can result in charge memory states with long retention times, relatively large extrema spacings, and analog tunability.

Fig. 1a schematically illustrates the shear exfoliation process for producing few-layer TMDC device structures. **Fig. 1b** shows the high-resolution TEM image of the cleaved surface of an as-exfoliated few-layer WSe₂ flake. This cleaved surface exhibits a quasi-periodic Moire pattern, which indicates the existence of exfoliation-induced interlayer deformation features (*e.g.*, interlayer twists or ripplocations) in the few-layer WSe₂ flake. Based on our density functional theory (DFT) simulation results, such interlayer deformation structures are expected to result in charge-trapping states with large binding energies and therefore long charge-retention times. Our FETs made from such exfoliated few-layer WSe₂ flakes exhibit long-lasting, metastable charge memory states with large extrema spacings and analog tunability, as demonstrated in **Fig. 1c**, which strongly supports the expectation from the DFT simulation. We further demonstrated that such multiple charge memory states have a good cycling durability, as demonstrated by the cycling endurance test result in **Fig. 2a**. Through investigating the temperature-dependent retention characteristics of few-layer WSe₂ FETs, we estimated the binding energies of holes and electrons in exfoliated few-layer WSe₂ flakes to be 0.7 and 0.4 eV, respectively. In the presentation, we will further present the neuromorphic signal processing results using our few-layer WSe₂ FETs with exfoliation-induced interlayer deformation structures.

This work advanced the critical nanofabrication and materials processing technologies for leveraging the unique charge-retention and mechanical properties of TMDC layers for new memory-related applications.

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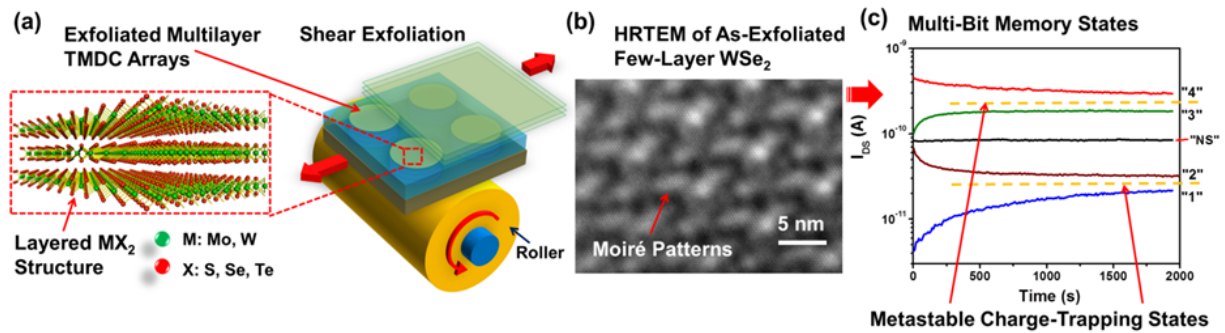


Fig. 1 (a) Schematic illustration of shear exfoliation, (b) High-resolution TEM image of the cleaved surface of a few-layer WSe₂ flake, (c) retention characteristics of a few-layer WSe₂ FET.

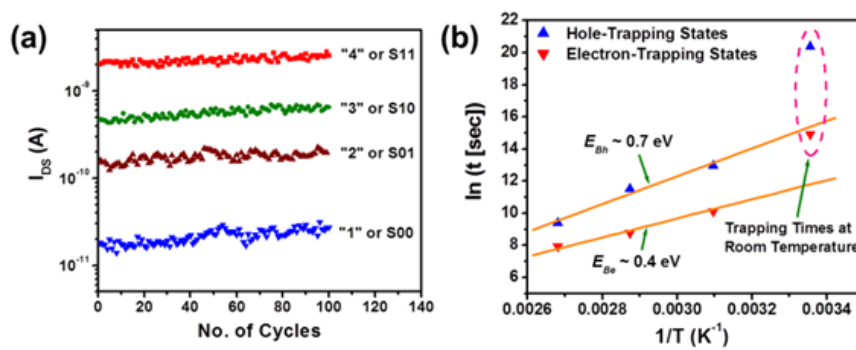


Fig. 2 (a) Cycling endurance test result of a 2-bit WSe₂ FET memory, (b) Trapping times (t) of hole-trapping and electron-trapping states in a few-layer WSe₂ FET, plotted as a function of temperatures, from which the binding energies for holes and electrons are extracted to be 0.7 and 0.4 eV, respectively.